

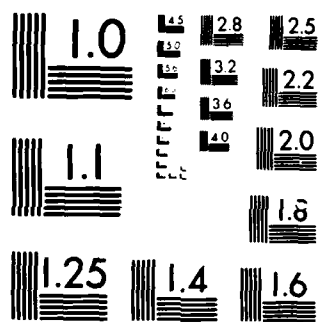
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ON K-BKZ AND OTHER VISCOELASTIC MODELS
AS CONTINUUM GENERALIZATIONS OF THE
CLASSICAL SPRING-DASHPOT MODELS

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December 1985

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ABSTRACT

An alternate constitutive formulation for visco-elastic materials, with particular emphasis on macromolecular viscoelastic fluids, is presented by generalizing Maxwell's idealized separation of elastic and relaxation mechanisms. The notion of relative rate of change of elastic stress is identified, abstracted, and formulated with the help of the established theory of finitely elastic isotropic materials. This gives a local rate type constitutive relation for an elastic mechanism in a simple material.

For the simplest class of viscoelastic polymer melts, the notion of rate of change of elastic stress and its damped accumulation is identified and formulated. Under conditions of moderate strain rates, this scheme implies the reliable K-BKZ model for a class of polymer melts. An obvious extension generalizes the remaining classical spring-dashpot models.

AMS (MOS) Subject Classification: 76A10

Key Words: Relative rate of change of elastic stress, instantaneous elasticity, configurational relaxation, material alignment, strain dependent elasticity modulus.

Work Unit Number 6 - Miscellaneous Topics

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Nomenclature

- (1) \mathcal{J} : The set of second order tensors. $A \in \mathcal{J}$ is identified with a 3x3 matrix in a Cartesian co-ordinate system.
- (2) \mathcal{J}^{sym} : The set of symmetric second order tensors.
- (3) \underline{Q} : A second order tensor \underline{Q} is orthogonal if and only if $\underline{Q}^T = \underline{Q}^{-1}$.
- (4) $\int_{t_0}^t \mathcal{H}(\underline{F}_{t_0}(\tau)) d\tau$: This is a symbol for the functional $\mathcal{H}: \mathcal{X} \rightarrow \mathcal{J}^{\text{sym}}$, where \mathcal{X} is the set of piecewise continuous and differentiable strain histories $\underline{F}_{t_0}: [t_0, t] \rightarrow \mathcal{J}$. Other functionals appearing in this paper, unless otherwise specified, should be interpreted in a similar manner.

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ON K-BKZ AND OTHER VISCOELASTIC MODELS AS CONTINUUM
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Amitabh Narain*

1. Introduction: The K-BKZ model ([2], [18], [33]) for viscoelastic fluids is known to give good experimental fit for rheometric data ([20], [33]) obtained for an identifiable class of melts. However, there are viscoelastic fluids and strain rates ([21], [38]) for which the K-BKZ model does not give good results. Among all the models for viscoelastic fluids, Maxwell's model ([28]) of a spring and a dashpot in series has always been used to give a qualitative meaning to elasticity and relaxation found in these fluids.

In this paper we show that if Maxwell's notions are quantified and generalized for an idealized *strictly viscoelastic fluid* possessing only the two mechanisms of fluid elasticity and configurational stress relaxation, then, at moderate strain rates, it leads to a K-BKZ model. The single integral models due to Doi-Edwards [13], Curtiss and Bird [12], Bernstein, Kearsley and Zapas [2], Rivlin and Sawyers [30] are closely related and, in a sense, more general than a generalized Maxwell model. A survey article of Tanner [33] and rheometric models of Wagner ([36], [37]) and Laun [20] make it worthwhile to understand the *scopes and limitations* of the K-BKZ model.

Most viscoelastic models fall in the category of Noll's simple materials [25] and Coleman and Noll's simple fluids [9]. Here we abstract and generalize the essential physical notion of Maxwell in

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such a way that the formulation is consistent with the statements of a Simple Material ([25], [34]), Objectivity ([25], [35]), and material symmetry ([25], [35]) associated with the natural rest states of a simple material.

The standard definition of a simple fluid is given by Coleman and Noll [9] in terms of a functional \mathcal{Y} such that the extra-stress is given by

$$\underline{T} + p\mathbf{1} = \mathcal{Y}_{s=0}^{\infty} [\underline{C}_t(t-s)]; \quad (1.1)$$

where \underline{T} is the Cauchy stress, p is the pressure, and $\underline{C}_t(t-s)$ is the right Cauchy-Green tensor evaluated at the past time $\tau = t-s \leq t$.

In order to model the simplest viscoelastic fluid within the framework of (1.1), the notion of *relative rate of change of elastic stress* is identified in the established theory of finitely elastic isotropic materials. This allows changes in stress on a deforming elastic element to be interpreted as a response to the non-dissipative changes occurring in the *material alignment* within the element. It is then noted that a change in *material alignment* can be important in a solid as well as in some fluids. However, a solid has a preferred material alignment in its rest state and a fluid has no preferred material alignment in its rest state. This fact is used in introducing and formulating the concept of *relative rate of change of elastic stress* $\dot{\underline{T}}_t(\tau)$ for an incompressible strictly viscoelastic fluid. A strictly viscoelastic fluid is a generalized Maxwell fluid endowed with the two mechanisms of elasticity and configurational relaxation. Under the conditions of moderate strain rates, we develop the following generalized Maxwell model:

$$\begin{aligned} \underline{T} &= -p \underline{1} + \int_0^\infty \tilde{G}(\sigma) \dot{\underline{C}}_t(t-\sigma) d\sigma, \quad t > 0. \\ \dot{\underline{C}}_t(\tau) &= \frac{d}{d\tau} [\phi(\underline{C}_t(\tau))], \\ \phi(\underline{C}_t(\tau)) &= [\phi_1(I, II) \underline{C}_t(\tau) + \phi_2(I, II) \underline{C}_t^{-1}(\tau)] \\ &\equiv -k[f_1(I, II) \underline{C}_t(\tau) + f_2(I, II) \underline{C}_t^{-1}(\tau)], \end{aligned} \quad (1.2)$$

$$I = \text{tr } \underline{C}_t(\tau),$$

and

$$II = \text{tr } \underline{C}_t^{-1}(\tau).$$

In (1.2), $\tilde{G}(0) = 1$, $\tilde{G}(\infty) = 0$, \tilde{G} is a non-dimensional monotonically decreasing relaxation function. The functions f_1 and f_2 are scalar valued and satisfy the conditions of zero extra stress for a rest history

$$f_1(3, 3) + f_2(3, 3) = 0,$$

and the condition for compatibility with linear viscoelastic fluids

$$\begin{aligned} \{f_2(3, 3) - f_1(3, 3)\} &+ \left\{ \frac{\partial f_1}{\partial II}(3, 3) - \frac{\partial f_1}{\partial I}(3, 3) \right\} \\ &+ \left\{ \frac{\partial f_2}{\partial II}(3, 3) - \frac{\partial f_2}{\partial I}(3, 3) \right\} = 1. \end{aligned}$$

Once a Maxwell model is generalized to (1.3), we present obvious generalizations of other spring dashpot models for Jeffrey's liquid (see [27]) and the standard linear viscoelastic solid ([14], p. 175).

2. The spring and dashpot models.

We will now recall some classical spring and dashpot models. For a long time it has been recognized (see [11], p. 26) that, if one does not take the analytical form of these models literally, the physical ideas underlying the classical mechanical models should have a valid qualitative generalization to general three dimensional straining.

a. A spring as a model for finite elasticity.

When a non-linear spring (see Fig. 2.1) has a strain $\epsilon(t)$ with respect to its unstretched configuration ($\epsilon(t_0)=0$) occupied at some time t_0 , the stress $\sigma(t)$ is given by a function g such that

$$\sigma(t) = g(\epsilon(t)); \text{ and } g(0) = 0. \quad (2.1)$$

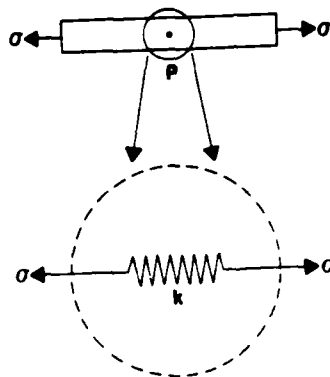


Fig. 2.1: A spring as a model of one dimensional elastic solid.

This constitutive assumption also can be written in the form

$$\sigma(t) = \int_{t_0}^t \dot{\pi}(\tau) d\tau,$$

where

$$\dot{\pi}(\tau) = \dot{\sigma}(\tau) = \frac{d}{d\tau} g(\epsilon(\tau)), \tau \in [t_0, t]. \quad (2.2)$$

This emphasizes the notion that this model has an undamped accumulation of elastic stress rate $\dot{\pi}(\tau)$. We wish to use the notion of

accumulation of elastic stress rate to develop a constitutive theory later in this paper.

b. Maxwell model of a spring and a dashpot in series.

The Maxwell model, with a spring and a dashpot in series ([27], p. 21), was meant to model the one dimensional straining of a viscoelastic fluid (see Figure 2.2).

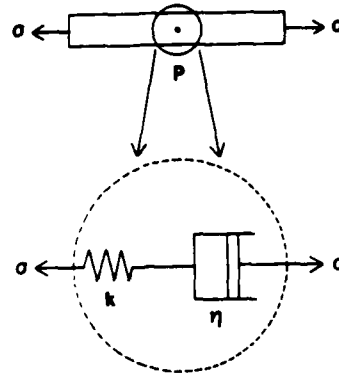


Fig. 2.2: A schematic representation of a Maxwell model. The mechanical devices within the circle represent physical mechanisms in a neighbourhood of the particle P.

If $\epsilon(t-s) - \epsilon(t)$ is the relative axial strain of a fluid element from the current strain $\epsilon(t)$, then in this model, the stress σ on the particle P at time t is given by

$$\begin{aligned}\sigma(P,t) &= \int_{-\infty}^t \exp(-(t-t')/\lambda) k \dot{\epsilon}(t') dt' \\ &= \int_0^{\infty} \exp(-s/\lambda) \{k \dot{\epsilon}(t-s)\} ds,\end{aligned}\tag{2.3}$$

where ϵ is the strain, and $\lambda = \frac{\eta}{k}$. In Fig. 2.2, k is the linear spring constant and η is the dashpot coefficient ([14], p. 175).

Eq. (2.3)₂ has the following alternate interpretation. Principal changes in stress occur through the elastic mechanism represented by the spring. The rate of change of elastic stress

$\dot{\pi}(t-s)$ (or $\dot{\pi}_t(t-s)$) at any past time $\tau = t-s$, in this case, is given by

$$\dot{\pi}(t-s) = k\dot{\epsilon}(t-s). \quad (2.4)$$

Because of the relaxation mechanism, the current value of stress can be interpreted as a damped and weighted accumulation of previous elastic changes in stress. The non-dimensional relaxation function $\exp(-s/\lambda)$ can be generalized to $\tilde{G}(s)$ and a Boltzmann form of the Maxwell model may be written in the form

$$\sigma(x,t) = \int_0^{\infty} \tilde{G}(s) \dot{\pi}(t-s) ds. \quad (2.5)$$

In (2.5), $\tilde{G}(s)$ is assumed to be a positive, monotonically decreasing, and piecewise continuously differentiable function such that

$$\tilde{G}(0) = 1,$$

and

$$\lim_{s \rightarrow \infty} \tilde{G}(s) = 0. \quad (2.6)$$

The above interpretation of the Maxwell model will provide some very interesting consequences when it is suitably generalized later in Sections 5, 6, and 7 of this paper.

c. The standard linear viscoelastic solid.

The standard linear viscoelastic solid ([14], p. 175) consists of a Maxwell element in parallel with a spring (see Fig. 2.3).

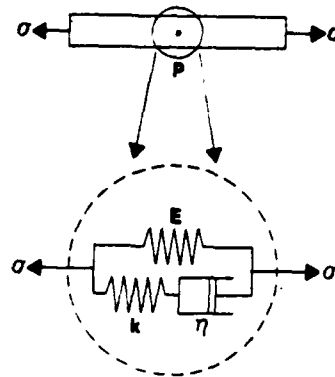


Fig 2.3: The spring in the above model provides the solid type behaviour. Viscoelastic mechanism comes through the Maxwell component of the model.

In this case, it is well known that σ in Fig. 2.3 is given by

$$\sigma(P,t) = E\varepsilon(t) + \int_0^{\infty} \exp(-s/\lambda) \{k\dot{\varepsilon}(t-s)\} ds. \quad (2.7)$$

d. Jeffrey's model with Newtonian viscosity.

To allow for noticeably distinct effects of rapid relaxation at very early times, presumably associated with Newtonian liquids, it is customary to allow for analogues of Jeffrey's liquid [27]. The mechanical model corresponds to a Maxwell element and a dashpot in parallel (see Fig. 2.4).

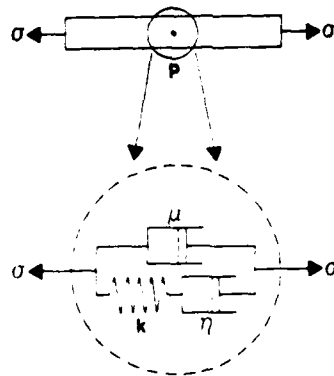


Fig. 2.4: The two dashpots have the coefficients of μ and η . The linear spring constant is k .

Here it follows that σ in Fig. 2.4 is given by

$$\sigma(P,t) = \mu \frac{d}{dt} (\varepsilon(\tau) - \varepsilon(t)) \Big|_{\tau=t} + \int_0^{\infty} \exp(-s/\lambda) \{k\dot{\varepsilon}(t-s)\} ds. \quad (2.8)$$

3. A brief review of the theory of simple materials.

Before we give a qualitative continuum generalization of the notions in section 2, it is convenient to briefly review the general scheme of Noll [25] and Coleman and Noll [9]. A standard single reference for such matters is Truesdell and Noll [35].

Let \underline{X} be the place occupied by a particle P of a simple material in its natural stress free configuration κ at time t_0 . Let the current position of the particle P at time t be \underline{x} , and let the motion for $\tau \in [t_0, t]$ be given by either $\underline{\xi} = \underline{x}_t(\underline{x}, \tau)$ or $\underline{\xi} = \underline{x}_{t_0}(\underline{X}, \tau)$. Clearly, at $\tau = t_0$, we must have

$$\underline{x} = \underline{x}_t(\underline{x}, t_0) = \underline{x}_{t_0}(\underline{X}, t_0), \quad (3.1)$$

which may be inverted to obtain

$$\underline{X} = \underline{x}_t^{-1}(\underline{x}, t_0), \quad (3.2)$$

and, thus

$$\underline{\xi} = \underline{x}_t(\underline{x}_t^{-1}(\underline{X}, t_0), \tau) = \underline{x}_{t_0}(\underline{X}, \tau).$$

The deformation gradients $\underline{F}_{t_0}(\underline{X}, t)$ and $\underline{F}_t(\underline{x}, t_0)$ are defined by

$$\underline{F}_{t_0}(\underline{X}, t) = \text{Grad } \underline{x}_{t_0}(\underline{X}, t),$$

and

$$(3.3)$$

$$\underline{F}_t(\underline{x}, t_0) = \text{grad } \underline{x}_t(\underline{x}, t_0),$$

and we note that

$$\underline{F}_t(\underline{x}, t_0) = \underline{F}_{t_0}(\underline{X}, t)^{-1}. \quad (3.4)$$

The Cauchy stress $\underline{T}(\underline{x}, t)$ of a homogeneous simple material for which the deformation is measured from its natural configuration is specified by a response functional $\kappa \mathcal{H}_1$, such that

$$\underline{T} = \kappa \mathcal{H}_1^t (F_{t_0}(\cdot)). \quad (3.5)$$

Since

$$\underline{F}_t(\underline{x}, \tau) \underline{F}_{t_0}(\underline{x}, t) = \underline{F}_{t_0}(\underline{x}, \tau), \quad (3.6)$$

it then follows that

$$\underline{T} = {}^{\kappa} \mathcal{H}_{\tau=t_0}^t (\underline{F}_t(\tau) \underline{F}_{t_0}(t)). \quad (3.7)$$

Note that the form of the functional ${}^{\kappa} \mathcal{H}_1$ depends on the natural configuration κ . For simple materials the restriction of Objectivity ([25], [34]) on ${}^{\kappa} \mathcal{H}_1$ is given by

$${}^{\kappa} \mathcal{H}_{\tau=t_0}^t (Q(\tau) \underline{F}_t(\tau) Q^T(t) \quad Q(t) \underline{F}_{t_0}(t)) = Q(t) {}^{\kappa} \mathcal{H}_{\tau=t_0}^t (\underline{F}_t(\tau) \underline{F}_{t_0}(t)) Q^T(t), \quad (3.8)$$

for any orthogonal tensor function $Q(\tau)$, $t_0 \leq \tau \leq t$. Using the polar decomposition [16]

$$\underline{F}_t(\tau) = \underline{R}_t(\tau) \underline{U}_t(\tau) = \underline{V}_t(\tau) \underline{R}_t(\tau) \quad (3.9)$$

and choosing

$$Q(\tau) = \underline{R}_t(\tau)^T, \quad (3.10)$$

we then see that (3.8) yields

$${}^{\kappa} \mathcal{H}_{\tau=t_0}^t (\underline{F}_t(\tau) \underline{F}_{t_0}(t)) = {}^{\kappa} \mathcal{H}_{\tau=t_0}^t (\underline{U}_t(\tau) \underline{F}_{t_0}(t)). \quad (3.11)$$

Thus, we have

$$\underline{T} = {}^{\kappa} \mathcal{H}_{\tau=t_0}^t (\underline{U}_t(\tau) \underline{F}_{t_0}(t)) \equiv {}^{\kappa} \mathcal{H}_{\tau=t_0}^t (\underline{U}_t(\tau), \underline{F}_{t_0}(t)). \quad (3.12)$$

The new response functional ${}^{\kappa} \mathcal{H}$ above is defined and introduced for convenience. Objectivity requires that ${}^{\kappa} \mathcal{H}$ satisfy

$$Q(t) {}^{\kappa} \mathcal{H}_{\tau=t_0}^t (\underline{U}_t(\tau), \underline{F}_{t_0}(t)) Q^T(t) = {}^{\kappa} \mathcal{H}_{\tau=t_0}^t (Q(t) \underline{U}_t(\tau) Q^T(t), Q(t) \underline{F}_{t_0}(t)) \quad (3.13)$$

for every orthogonal Q . The form of (3.12) emphasizes that only the intermediate local shape history with respect to the current

configuration (measured through $\underline{U}_t(\tau)$, $t_0 \leq \tau \leq t$) and the current local shape with respect to the reference configuration κ (measured through $\underline{F}_{t_0}(t)$) affect the current value of the Cauchy stress at a material particle. Now the particular natural rest configuration κ in the development above may be indistinguishable from others in a set of natural configurations which are related to κ by transformations belonging to a *symmetry group* [16] associated with the local material alignment. A common statement of this symmetry invariance requirement states that (c.f., [34]),

$$\begin{aligned} \underline{T} &= \bigwedge_{\tau=t_0}^t (\underline{U}_t(\tau), \underline{F}_{t_0}(t) \underline{H}) \\ &= \bigwedge_{\tau=t_0}^t (\underline{U}_t(\tau), \underline{F}_{t_0}(t)) \end{aligned} \quad (3.14)$$

for every tensor \underline{H} in the symmetry group \mathcal{S}_κ associated with κ for a given material. For an isotropic solid, \mathcal{S}_κ is the full orthogonal group ([34]), and it is well known that in this case (3.9) and (3.14) may be combined (with $\underline{H} = \underline{R}_{t_0}(t)^T$) to give

$$\underline{T} = \bigwedge_{\tau=t_0}^t (\underline{U}_t(\tau), \underline{V}_{t_0}(t)). \quad (3.15)$$

One finds, using the uniqueness of the decomposition in (3.9), that

$$\underline{V}_{t_0}(t)^{-1} = \underline{U}_t(t_0). \quad (3.16)$$

It is also known that the right Cauchy - Green tensor $\underline{C}_t(\tau)$ is given by

$$\underline{C}_t(\tau) = \underline{U}_t(\tau) \underline{U}_t(\tau)^T, \tau \in [t_0, t]. \quad (3.17)$$

Using (3.16) and (3.17) in (3.15), we introduce a functional *key* such that

$$\underline{T} = \kappa \int_{\tau=t_0}^t [\underline{C}_t(\tau), \underline{C}_t(t_0)]. \quad (3.18)$$

In (3.18), the material is in its undistorted configuration up to time t_0 (i.e., $\dot{\underline{C}}_t(\tau) = 0$ for $\tau \leq t_0$). This means that the representation of the functional in (3.18) does not depend on the specific choice of the undistorted configuration κ and the associated time t_0 (see Appendix A). This allows us to assert that the natural state of an isotropic simple solid determines a response functional $\mathcal{Y}_{\underline{C}_t(t_0)}$ such that

$$\underline{T} = \int_{\tau=t_0}^t \mathcal{Y}_{\underline{C}_t(t_0)}[\underline{C}_t(\tau)]. \quad (3.19)$$

In (3.19), the explicit dependence of $\mathcal{Y}_{\underline{C}_t(t_0)}$ on $\underline{C}_t(t_0)$ accounts for the differences in *material alignment* of the current state and the preferred isotropic *material alignment* of the rest state. Furthermore, the Objectivity requirement (3.13) reduces to

$$\mathcal{Y}_{\underline{Q}\underline{C}_t(t_0)\underline{Q}^T}[\underline{Q}\underline{C}_t(\tau)\underline{Q}^T] = \underline{Q} [\mathcal{Y}_{\underline{C}_t(t_0)}(\underline{C}_t(\tau))]\underline{Q}^T \quad (3.20)$$

for every orthogonal tensor \underline{Q} .

For an incompressible ([34], p. 44) simple fluid, \mathcal{R}_κ is the full unimodular group (i.e., any $\underline{H} \in \mathcal{R}_\kappa$ satisfies $\det \underline{H} = 1$) and the resulting constitutive theory reduces to (see [34], p. 57)

$$\underline{T} = -p\underline{1} + \int_{\tau=t_0}^t [\underline{C}_t(\tau)], \quad (3.21)$$

with the constraint

$$\det \underline{C}_t(\tau) = 1, \quad \tau \in [t_0, t].$$

In (3.21), p is a constitutively indeterminate pressure and \mathcal{Y} is independent of the choice of reference configuration. Also, (3.21) allows the possibility of finding an undistorted state only as the time $t_0 \rightarrow -\infty$ for a given $0 < t < \infty$. However, if t_0 is finite

($t_0 > -\infty$), then the statement (A.12) in Appendix A allows us to set $t_0 = -\infty$ without changing the value of the stress. Therefore, without loss of generality, we can set $t_0 = -\infty$ in (3.21) and this makes (3.21) equivalent to the statement in (1.1).

4. Finite elasticity and the notion of relative rate of change of elastic stress $\dot{\underline{T}}_t(\tau)$.

Let us consider an isotropic solid which is purely elastic and therefore is an analogue of the spring in section 2a. In this case (3.18) is given by

$$\begin{aligned}\underline{T}(\underline{x}, t) &= \kappa \int_{\tau=t_0}^t [\underline{C}_t(\tau), \underline{C}_t(t_0)] \\ &= f^K(\underline{C}_t(t_0)) \\ &\stackrel{\text{def}}{=} g^K(\underline{B}(t)),\end{aligned}\tag{4.1}$$

where

$$\begin{aligned}\underline{B}(t) &\stackrel{\text{def}}{=} \underline{F}_{t_0}(t) \underline{F}_{t_0}(t)^T \\ &= \underline{C}_t(t_0)^{-1}.\end{aligned}\tag{4.2}$$

and

$$\underline{Q} g^K(\underline{B}(t)) \underline{Q}^T = g^K(\underline{Q} \underline{B}(t) \underline{Q}^T)$$

for every orthogonal \underline{Q} . Similarly, the intermediate stress $\underline{T}(\underline{\xi}, \tau)$ at a time $\tau \in [t_0, t]$, is given by (see Appendix B):

$$\begin{aligned}\underline{T}(\underline{\xi}, \tau) &= f^K(\underline{C}_\tau(t_0)), \\ &= \phi_{\underline{C}_t(t_0)}(\underline{C}_t(\tau)),\end{aligned}\tag{4.3}$$

where the response function $\phi_{\underline{C}_t(t_0)}$ depends explicitly on $\underline{C}_t(t_0)$.

The restriction of Objectivity (see (B.5) in Appendix B) becomes

$$\underline{Q} \underline{C}_t(t_0) \underline{Q}^T (\underline{Q} \underline{C}_t(\tau) \underline{Q}^T) = \underline{Q} [\phi_{\underline{C}_t(t_0)}(\underline{C}_t(\tau))] \underline{Q}^T\tag{4.4}$$

for every orthogonal tensor \underline{Q} and $\tau \in [t_0, t]$.

We define the material derivative of (4.3) at time τ as rate of change of elastic stress $\dot{\underline{T}}_t(\tau)$ and write

$$\begin{aligned}\dot{\underline{T}}(\underline{x}, \tau) &= \frac{d}{d\tau} [\phi_{\underline{C}_t(t_0)}(\underline{C}_t(\tau))] \\ &= \dot{\underline{\pi}}_t(\tau).\end{aligned}\quad (4.5)$$

Since at time t_0 , the particle is at \underline{x} and the material element is in its natural configuration κ , we have

$$\underline{T}(\underline{x}, t_0) = \underline{0}.\quad (4.6)$$

Collecting (4.5), (4.6), and (B.3)₃ from Appendix B, we have

$$\begin{aligned}\underline{T}(\underline{x}, t) &= \int_{t_0}^t \dot{\underline{\pi}}_t(\tau) d\tau \\ &= \int_{t_0}^t \frac{d}{d\tau} [\phi_{\underline{C}_t(t_0)}(\underline{C}_t(\tau))] d\tau \\ &= \phi_{\underline{C}_t(t_0)}(\underline{1}) = g^{\kappa}(\underline{B}(t)).\end{aligned}\quad (4.7)$$

The first equality in (4.7)₁ allows us to interpret the current value of Cauchy stress as an undamped accumulation of elastic stress rate $\dot{\underline{\pi}}_t(\tau)$. This stress rate measures the rate of non-dissipative changes occurring in the *material alignment* of the element. We wish to borrow this notion of *instantaneous elasticity* or *relative rate of change of elastic stress* $\dot{\underline{\pi}}_t(\tau)$ for modeling instantaneous elasticity in fluids. Of course, for a fluid, if

$$\dot{\underline{\pi}}_t(\tau) = \frac{d}{d\tau} [\phi_{\underline{C}_t(t_0)}(\underline{C}_t(\tau))], \tau \in [t_0, t],\quad (4.8)$$

then the additional restriction of fluid symmetry leading to (3.20) suggests that $\phi_{\underline{C}_t(t_0)}$ in (4.8) will become independent of $\underline{C}_t(t_0)$. This is because the rest state of a fluid has no preferred *material alignment* but there are differences in *material alignment* between two unrelaxed stressed states of an elastic fluid element.

It is further noted that it is impossible to have an incompressible purely elastic fluid in the sense of an elastic

solid in (4.7). This is because an incompressible purely elastic fluid will have

$$\begin{aligned} \underline{T}(\underline{x}, t) &= \int_{t_0}^t \dot{\underline{T}}_t(\tau) d\tau \\ &= \underline{\phi}(\underline{1}) - \underline{\phi}(\underline{C}_t(t_0)), \end{aligned} \quad (4.9)$$

$$\text{for } \dot{\underline{T}}_t(\tau) = \frac{d}{d\tau} [\underline{\phi}(\underline{C}_t(\tau))].$$

Now, for a constant $\underline{C}_t(t_0) \neq \underline{1}$ and $t \rightarrow \infty$, $\underline{T}(\underline{x}, t)$ in (4.9) does not tend to a hydrostatic pressure for an arbitrary isotropic $\underline{\phi}$. This is in contradiction to the notion of a rest state for a fluid.

However, as we see in the next sections, a fluid (like Maxwell model) can possess *instantaneous elasticity* given by (4.9)₂ if it has some additional mechanism of stress relaxation.

5. On phenomenological motivation for modeling a class of polymer melts as a generalized Maxwell model.

We now wish to motivate the suggested notion of rate of change of elastic stress. For this purpose, we consider a class of polymer melts as a representative strictly viscoelastic fluid. Therefore, in what follows, the following specific phenomenological picture of macromolecular deformation is essential to the validity of the modeling presented in this paper.

Here we are concerned with polymer melts which contain numerous supple (flexible) tubelike strands of macromolecules and their networks. During deformation, an obvious example being extensional flows, an arbitrary material element of the fluid may contain somewhat oriented strands of macromolecules. Yet the stress free *prolonged rest state* corresponds to tensionless strands of macromolecules which are randomly oriented within a specified fluid element. This random orientation of inert tensionless macromolecules is required for consistency with the *unimodular symmetry* [34] expected from the prolonged rest state of any simple fluid. However, when a stressed fluid element is deformed from one shape to another, the supple resilient macromolecules conform to the new shape and some stress is generated through tension in the macromolecules. As the *fluid element* is made to change its shape by incremental stress, this new deformed shape has some elastic propensity to return to its original shape. Accompanying this momentary non-dissipative change in the alignment of the macromolecules, on a lagging time scale, there is a release of tension through dientanglement and readjustment of macromolecular networks. This

release of tension *within* an oriented shape of macromolecules is assumed here to be the principle mechanism of *configurational relaxation*. This notion is perhaps the same as the notion of *configurational relaxation* proposed by Giesekus [15]. Of course, if the fluid element stops deforming, this *configurational relaxation* will eventually produce a state of tensionlessness and this state will easily allow randomizing of macromolecular orientation to take place. Therefore, any randomizing associated with relaxation is assumed to be noticeable only in cessational flows (straining of a fluid element stops), and even in this case *configurational relaxation* is assumed to be the dominant part of relaxation. Therefore, we assume that changes in stress are due to two mechanisms: one is through an elastic mechanism and the other is through a relaxation mechanism. The elastic mechanism corresponds to resilient alignment of supple macromolecules within the changing shape of a fluid element. We further assume, despite the presence of some possible reptations [13], that all effects of material alignment can be modeled by the changing shape of a fluid element. Since we are interested in the current value of the Cauchy stress, it is beneficial to use the current configuration of a fluid element for modeling different elastic contributions from differently aligned macromolecules in shapes different than the current one. For this class of polymer melts, as said earlier, we assume that the effects of the tendency of macromolecules to randomize within a deforming material element is insignificant when compared to *configurational relaxation* and the elastic mechanism associated with direct mechanical straining*. This does not mean that all

*The fact that randomization in some melts and concentrated solutions can be ignored is also discussed by Marrucci (Polymers, Liquid Crystals, and Low Dimensional Solids, Plenum Press, p. 149, 1984).

polymers can be modeled by ignoring effects of macromolecular randomization within a given shape of a material element. In fact, the known theories ([5], [6]) of rubber elasticity and other theories of entropic elasticity ([1], [6]), can possibly be adapted into this proposed modeling approach to include the effects of macromolecular randomization. However, in this paper, we restrict ourselves to those melts which have ignorable contribution associated with randomization.

Therefore, when a strictly viscoelastic element (identified at the current time t) is strained from a configuration $C_t(\sigma + \Delta\sigma) \stackrel{\text{def}}{=} C_t(t - (\sigma + \Delta\sigma))$ to a configuration $C_t(\sigma)$, the change in shape activates a direct change in stress (modeled as elastic change) $\pi_t^{\Delta\sigma} = \dot{\pi}_t(t - \sigma)\Delta\sigma + o(\Delta\sigma)$ near the past time $\tau = t - \sigma$. Having looked at the notion of *relative rate of change of elastic stress* in section 4, we borrow the constitutive model (4.8) for dependence of $\dot{\pi}_t(t - \sigma)$ on the strain $C_t(t - \sigma)$ and on the strain rate $\dot{C}_t(t - \sigma)$.

Because of the presence of relaxation mechanism, the current value of Cauchy stress $T(x, t)$ is a damped accumulation of incremental stresses $\pi_t^{\Delta\sigma}$. This notion is a qualitative generalization of Maxwell model and is formally stated in the next section.

6. On idealized presence of elastic and dissipative mechanisms for a generalized Maxwell model.

For a strictly viscoelastic fluid, we assume the standard kinematics of section 3. In addition, at any instant of time t , while chasing an identifiable neighbourhood of a particle P , there exists a rate of change of elastic stress history

$$\dot{\tilde{\pi}}(t): [0, \infty) \longrightarrow \mathcal{J}^{\text{sym}}, \quad (6.1)$$

and $\dot{\tilde{\pi}}(t)(s) = \dot{\tilde{\pi}}(P, t-s) = \dot{\tilde{\pi}}_t(x, t; t-s)$ is the relative rate of change of elastic stress. For $\tau \in [t_0, t]$ and $t_0 \geq -\infty$, (4.8) is written as

$$\begin{aligned} \dot{\tilde{\pi}}_t(\tau) &= \frac{d}{d\tau} [\phi_{C_t(t_0)}(C_t(\tau))] \\ &= D\phi_{C_t(t_0)}(C_t(\tau)) \dot{C}_t(\tau). \end{aligned} \quad (6.2)$$

In (6.2), the strain dependent elasticity modulus " $D\phi_{C_t(t_0)}(C_t(\tau))$ " models the effect of material alignment in changing shapes and the strain rate $\dot{C}_t(\tau)$ is the principle cause of changes in stress.

For now, we postulate $\dot{\tilde{\pi}}_t(\tau)$ to be given by (6.2) subject to further restrictions imposed by the increased symmetry of the rest state of a fluid. To model the effect of relaxation and its possible coupling with elasticity, we postulate that the current value of extra stress for an incompressible generalized Maxwell fluid is given by

$$\tilde{T}(x, t) = -p\tilde{1} + \mathcal{R}_{\tau=t_0}^t [\dot{\tilde{\pi}}_t(\tau)], \quad (6.3)$$

where \mathcal{R} is an accumulation functional associated with relaxation. without loss of generality, the rest state is taken to be at $t_0 = -\infty$ and (6.3) is rewritten as

$$\underline{T} = -p\underline{1} + \int_{-\infty}^t [\dot{\underline{\pi}}(t-s)] \stackrel{\text{def}}{=} -p\underline{1} + \underline{R}[\dot{\underline{\pi}}(t)]. \quad (6.4)$$

On substituting (6.2) in (6.4) and then applying the notion of fluid symmetry leading to (3.21), one can show that

$$\dot{\underline{\pi}}(t) = \frac{d}{dt} [\phi(\underline{C}_t(t))], \quad (6.5)$$

for $t \in (-\infty, t]$. Clearly (6.4) and (6.5) together give a special class of simple fluid given by (1.1). The mathematical assumptions on \mathcal{U} which leads to (6.4) and (6.5) may be an interesting area of investigation. However, in this paper, as is the case with the original Maxwell model, the notion of elasticity and relaxation are modeled directly on the basis of the phenomenology described and assumed in section 5.

We now see that the restriction of Objectivity in (4.4) and (4.8) implies isotropy of the function ϕ in (6.5). Therefore

$$\phi(\underline{Q} \underline{C}_t(t) \underline{Q}^T) = \underline{Q} \phi(\underline{C}_t(t)) \underline{Q}^T \quad (6.6)$$

for any orthogonal \underline{Q} . It is noted that the restriction (6.6) is imposed by the fact that our Euclidean space has no preferred directions and it has nothing to do with material isotropy at the strain $\underline{C}_t(t)$. In fact, we recall:

$$\hat{\underline{C}}(\underline{C}_t(t-s)) = \underline{D}\phi(\underline{C}_t(t-s)) \quad (6.7)$$

in (6.2) measures the effects of relative differences in material alignments between the continuum configurations at times $\tau = t-s$ and $\tau = t$. It is instructive to relate the choice in (6.5) to the notion of elastic resilience alluded to in the phenomenological description of section 5. For this, consider a past time $\tau = t-(t+\Delta t)$ and the strain $\underline{C}_t(t-(t+\Delta t)) \stackrel{\text{def}}{=} \underline{C}_t(t)(t+\Delta t)$. When the fluid element

is brought to the configuration $C_t(\sigma)$, we do so by an incremental stress

$$\begin{aligned}\pi_t^{\Delta\sigma} &= \int_{t-(\sigma+\Delta\sigma)}^{t-\sigma} \dot{\pi}_t(t-s) ds \\ &= [\phi(C_t(t-\sigma)) - \phi(C_t(t-(\sigma+\Delta\sigma)))] \\ &= \phi(C_t(\sigma)) - \phi(C_t(\sigma+\Delta\sigma)).\end{aligned}\quad (6.8)$$

Now before relaxation sets in, if the incremental stress $\pi_t^{\Delta\sigma}$ was set to zero, (6.8) requires that we must have

$$\phi(C_t(\sigma+\Delta\sigma)) - \phi(C_t(\sigma)) = 0. \quad (6.9)$$

In the thought experiment resulting in (6.9), we find that if ϕ is invertible at $C_t(\sigma)$ (that is the elastic modulus $D\phi(C_t(\sigma))$ is such that $D\phi(C_t(\sigma))\underline{A} = 0$ implies $\underline{A} = 0 \in \mathcal{J}^{sym}$), then (6.9) implies

$$C_t(\sigma+\Delta\sigma) = C_t(\sigma). \quad (6.10)$$

We view (6.10) to be in accord with the physical notion of elastic recoil (see [4]) when the stress $\pi_t^{\Delta\sigma}$ is removed prior to the onset of relaxation.

Furthermore, if the maximum magnitude of strain $\tau \leftarrow \sup_{(-\infty, t]} |C_t(\tau) - \underline{1}|$ exists and is close to zero, then $\dot{\pi}_t(t-s)$ in (6.7) can be approximated by

$$\dot{\pi}_t(s) \approx \underline{\mathbb{C}} \dot{C}_t(t-s), \quad (6.11)$$

where $\underline{\mathbb{C}} = D\phi(\underline{1})$ is a constant fourth order tensor in the space of linear maps from \mathcal{J}^{sym} to \mathcal{J}^{sym} . The constant tensor $\underline{\mathbb{C}}$ in (6.11) makes sense because now there is no significant difference in material alignments at all times $\tau \leq t$. On applying the isotropy condition (6.2), we find that

$$\underline{Q} [\underline{\mathbb{C}} \dot{C}_t(t-s)] \underline{Q}^T = \underline{\mathbb{C}} [\underline{Q} \dot{C}_t(t-s) \underline{Q}^T] \quad (6.12)$$

for any orthogonal tensor \underline{Q} . Using a well known result [32] often

used in linear theory of elasticity, it follows that (6.12) implies that $\underline{\underline{C}}_t(t-s)$ must be of the form

$$\underline{\underline{C}}_t(t-s) = k \underline{\underline{C}}_t(t-s) + \nu \text{tr}(\underline{\underline{C}}_t(t-s)) \underline{\underline{1}} \quad (6.13)$$

for some constants k and ν . Eq. (6.12) and (6.13) embody the notion of a linear elastic mechanism and correspond to a linear spring in the Maxwell model of Fig. 2.2. The general isotropy condition in (6.6) along with an adaptation of theorems of Rivlin and Ericksen [29], and Serrin [32] implies that ϕ has a representation

$$\underline{\underline{\phi}}(\underline{\underline{C}}_t(t-\sigma)) = \phi_0(I, II) \underline{\underline{1}} + \phi_1(I, II) \underline{\underline{C}}_t(t-\sigma) + \phi_2(I, II) \underline{\underline{C}}_t^{-1}(t-\sigma),$$

where

(6.14)

$$I = \text{tr}(\underline{\underline{C}}_t(t-\sigma)), \text{ and } II = \text{tr}(\underline{\underline{C}}_t^{-1}(t-\sigma)).$$

As a result of the incompressibility condition (3.21)₂, the third invariant of $\underline{\underline{C}}_t(t-\sigma)$ does not appear in (6.14). ϕ_0 , ϕ_1 , and ϕ_2 in eq. (6.14) are material functions of the invariants I and II .

tion h be small. Of course we assume that the norm of $D\phi$ remains bounded in the space of linear maps from \mathcal{U}^{sym} to \mathcal{U}^{sym} . Therefore, this assumption allows for large and even unbounded magnitudes of strains as long as $\dot{C}_t(t-s)$ is small in the recent past. We now make an additional physical assumption regarding \tilde{G} . Let \tilde{G} be positive on $[0, \infty)$, piecewise continuously differentiable on $(0, \infty)$, monotonically decreasing on $[0, \infty)$,

$$\tilde{G}(0) = 1,$$

and

(7.5)

$$\tilde{G}(s) \rightarrow 0 \text{ as } s \rightarrow \infty$$

sufficiently fast for a meaningful integral in (7.3). The assumption (7.5) on \tilde{G} in (7.3) can be shown to follow from requiring that the magnitude of the traction vector on the boundary of a fluid element must never increase if the element was made to experience a static continuation (see [26] for the meaning of this term).

We note that $\dot{\pi}(t)$ has the physical dimensions ($ML^{-1}T^{-3}$) of rate of change of stress. This implies that $\tilde{G}(s)$ is nondimensional. This, in turn, suggests a provable implication that there exists at least one relaxation time $\bar{\lambda}$ and a function \bar{G} such that

$$\tilde{G}(s) = \bar{G}(s/\bar{\lambda}) \quad (7.6)$$

for every $s \geq 0$.

If we now consider histories $C(t)$ which are such that both $\|C(t) - I\|_h$ and $\|\dot{C}(t)\|_h$ are small in the inner product space induced by (7.1), then, the linearization (7.3) should be expected to reduce to the standard and general result of Coleman and Noll in (5.18) of [10]. For such a history we use the linear relation for $\dot{\pi}(t)$ in (6.13) and substitute it in (7.3).

After absorbing the multiples of unit tensor in the indeterminate pressure, we find

$$\underline{\tau}^{\text{Lin}} = -p_1 + \int_0^\infty \tilde{G}(s) k \dot{\underline{C}}_t(t-s) ds \quad (7.7)$$

The result in (7.7) is compatible with the standard linearization (cf. (5.18) of [10])

$$\underline{\tau}^{\text{Lin}} = -p_1 + \int_0^\infty G'(s) \underline{C}_t(t-s) ds. \quad (7.8)$$

The function \tilde{G} and G in (7.7) and (7.8) are related by

$$G(0) = k > 0,$$

and

$$G(s) \stackrel{\text{def}}{=} k \tilde{G}(s), \quad s \geq 0. \quad (7.9)$$

The result in (7.9) gives an important indication that the postulate in (6.3) is a good one. This is because $\sqrt{G(0)/\rho}$ is known to give the shock speed ([8], [23]) for a linearized strictly viscoelastic fluid given by (7.8). Interpretation of $G(0)=k$ as a modulus of the elastic mechanism in the fluid seems to give meaning to this result. The shock speed is therefore due to elastic resilience to impact. Also various results of Coleman, Gurtin and Herrera [9], Narain and Joseph [23], and Renardy [28] show that the amplitude of a shock decays with time as $\exp(tG'(0)/G(0))$. In the above interpretation, the decay is governed only by the parameter $\tilde{G}'(0) = G'(0)/G(0) < 0$, and this parameter is related only to the relaxation mechanism of a linearized strictly viscoelastic fluid. Similar interpretation of other formulas of Narain and Joseph [23] is discussed in Narain [22].

8. The assumptions underlying the K-BKZ type models for polymer melts.

In a survey article, Tanner [33] finds that the K-BKZ type model is superior regarding compatibility with experiments in a large class of motions of polymeric fluids. Tanner [33] indicated that there is no good qualitative understanding for this model. It is hoped that this paper contributes towards this purpose. The representation of the K-BKZ model seems to be based on the assumption that the fluid can be modeled as a generalized Maxwell model and that the strain-rates are moderate enough to ignore the second term in (7.3)₁. On substituting (6.5) in (7.3)₁, and ignoring the smaller term, one finds

$$\underline{\underline{T}} = -p\underline{\underline{1}} + \int_0^\infty -\tilde{G}(s) \frac{d}{ds} \phi(\underline{\underline{C}}_t(t-s)) ds. \quad (8.1)$$

The assumption of moderate strain rates seems to be justified by the fact that the K-BKZ model (8.1) is not accurate (see [21]) under high strain rates involved in experiments with multiple steps in strain. This is because, under these conditions, $||\dot{\underline{\underline{C}}}_t||_h$ and hence $o(||\dot{\underline{\underline{\pi}}}_t||_h)$ in (7.3)₁ is not negligible. Physically, this amounts to saying that the time scale associated with rate of strain is small compared to a characteristic time scale $\bar{\lambda}$ of relaxation (high Weissenberg Number, $\bar{\lambda} \frac{U}{L}$: see (3.15) of [11]). That is, rate of elastic energy build up far exceeds the dissipation rate and therefore the assumption of independent elastic and relaxation mechanisms in (8.1) is not very good.

On substituting (6.14) in (8.1) and integrating it by parts while absorbing the multiples of unit tensor $\underline{\underline{1}}$ in the pressure term, we find

$$\underline{T} = -p\underline{1} + \int_0^\infty \underline{\tilde{G}}'(\sigma) \{ \phi_1(I, II) \underline{C}_t(t-\sigma) + \phi_2(I, II) \underline{C}_t^{-1}(t-\sigma) \} d\sigma. \quad (8.2)$$

One can relate (8.2) to the popular K-BKZ form ([33]), by introducing

$$a(\sigma) \stackrel{\text{def}}{=} -k \tilde{G}'(\sigma) = -G'(\sigma), \sigma \geq 0$$

$$\frac{1}{k} \phi_1(I, II) \stackrel{\text{def}}{=} -f_1(I, II),$$

and

$$\frac{1}{k} \phi_2(I, II) \stackrel{\text{def}}{=} -f_2(I, II),$$

(8.3)

where $k > 0$ is the same as in (7.7).

Using (8.3), (8.2) is written in the form

$$\underline{T} = -p\underline{1} + \int_0^\infty a(\sigma) \{ f_1(I, II) \underline{C}_t(t-\sigma) + f_2(I, II) \underline{C}_t^{-1}(t-\sigma) \} d\sigma. \quad (8.4)$$

The functions f_1 and f_2 in (8.4) can be normalized by requiring the extra-stress to be zero for the rest history $\underline{C}_t(\tau) = \underline{1}$, $\tau \leq t$.

This gives

$$f_1(3, 3) + f_2(3, 3) = 0. \quad (8.5)$$

Furthermore, a linearization of (8.4) gives

$$\underline{T} = -p\underline{1} + \int_0^\infty a(\sigma) \hat{C} \underline{C}_t(t-\sigma) d\sigma, \quad (8.6)$$

where

$$\begin{aligned} \hat{C} = & \left\{ \frac{\partial f_1}{\partial I} (3, 3) - \frac{\partial f_1}{\partial II} (3, 3) \right\} + \left\{ \frac{\partial f_2}{\partial I} (3, 3) - \frac{\partial f_2}{\partial II} (3, 3) \right\} \\ & + \{ f_1(3, 3) - f_2(3, 3) \}. \end{aligned}$$

For (8.6) to be compatible with (7.8), it is required that

$$\hat{C} = -1. \quad (8.7)$$

8. The assumptions underlying the K-BKZ type models for polymer melts.

In a survey article, Tanner [33] finds that the K-BKZ type model is superior regarding compatibility with experiments in a large class of motions of polymeric fluids. Tanner [33] indicated that there is no good qualitative understanding for this model. It is hoped that this paper contributes towards this purpose. The representation of the K-BKZ model seems to be based on the assumption that the fluid can be modeled as a generalized Maxwell model and that the strain-rates are moderate enough to ignore the second term in (7.3)₁. On substituting (6.5) in (7.3)₁, and ignoring the smaller term, one finds

$$\underline{T} = -p\underline{1} + \int_0^\infty -\tilde{G}(s) \frac{d}{ds} \phi(\underline{C}_t(t-s)) ds. \quad (8.1)$$

The assumption of moderate strain rates seems to be justified by the fact that the K-BKZ model (8.1) is not accurate (see [21]) under high strain rates involved in experiments with multiple steps in strain. This is because, under these conditions, $||\dot{\underline{C}}_t||_H$ and hence $o(||\dot{\underline{\pi}}_t||_H)$ in (7.3)₁ is not negligible. Physically, this amounts to saying that the time scale associated with rate of strain is small compared to a characteristic time scale $\bar{\lambda}$ of relaxation (high Weissenberg Number, $\bar{\lambda} \frac{U}{L}$: see (3.15) of [11]). That is, rate of elastic energy build up far exceeds the dissipation rate and therefore the assumption of independent elastic and relaxation mechanisms in (8.1) is not very good.

On substituting (6.14) in (8.1) and integrating it by parts while absorbing the multiples of unit tensor $\underline{1}$ in the pressure term, we find

$$\underline{T} = -p\underline{1} + \int_0^\infty \underline{\tilde{G}}'(\sigma) \{ \phi_1(I, II) \underline{C}_t(t-\sigma) + \phi_2(I, II) \underline{C}_t^{-1}(t-\sigma) \} d\sigma. \quad (8.2)$$

One can relate (8.2) to the popular K-BKZ form ([33]), by introducing

$$a(\sigma) \stackrel{\text{def}}{=} -k \underline{\tilde{G}}'(\sigma) = -G'(\sigma), \sigma \geq 0$$

$$\frac{1}{k} \phi_1(I, II) \stackrel{\text{def}}{=} -f_1(I, II),$$

and

$$\frac{1}{k} \phi_2(I, II) \stackrel{\text{def}}{=} -f_2(I, II),$$

(8.3)

where $k > 0$ is the same as in (7.7).

Using (8.3), (8.2) is written in the form

$$\underline{T} = -p\underline{1} + \int_0^\infty a(\sigma) \{ f_1(I, II) \underline{C}_t(t-\sigma) + f_2(I, II) \underline{C}_t^{-1}(t-\sigma) \} d\sigma. \quad (8.4)$$

The functions f_1 and f_2 in (8.4) can be normalized by requiring the extra-stress to be zero for the rest history $\underline{C}_t(\tau) = \underline{1}, \tau \leq t$.

This gives

$$f_1(3, 3) + f_2(3, 3) = 0. \quad (8.5)$$

Furthermore, a linearization of (8.4) gives

$$\underline{T} = -p\underline{1} + \int_0^\infty a(\sigma) \hat{C} \underline{C}_t(t-\sigma) d\sigma, \quad (8.6)$$

where

$$\begin{aligned} \hat{C} = & \left\{ \frac{\partial f_1}{\partial I} (3, 3) - \frac{\partial f_1}{\partial II} (3, 3) \right\} + \left\{ \frac{\partial f_2}{\partial I} (3, 3) - \frac{\partial f_2}{\partial II} (3, 3) \right\} \\ & + \{ f_1(3, 3) - f_2(3, 3) \}. \end{aligned}$$

For (8.6) to be compatible with (7.8), it is required that

$$\hat{C} = -1. \quad (8.7)$$

The suitability of the above K-BKZ type model can only be verified by applications and experiments. Important semi-empirical formulations of Wagner ([36], [37]), Laun [20], and Chang and Lodge [7] can easily be shown to lie in the above framework with $f_1(I, II)=0$.

Since Lodge's rubberlike fluid [3] is a special case of (8.4), various experimental comparisons [33] suggest that it is advisable to do melt rheometry in this more general framework. The most important experimental verification and suggestion of the model in (8.4) can be found in Laun's paper [20]. This is briefly reinterpreted in section 11.

9. On the generalization of standard linear viscoelastic solid.

In this section we generalize the notion of a standard linear viscoelastic solid shown in Fig. 2.3.

For an isotropic viscoelastic solid, the development of section 3 leading to (3.14) holds. Using $\underline{B}(t)$ in (4.1)₄ and the fact that

$$\underline{B}(t) = \underline{V}_{t_0}(t) \underline{V}_{t_0}(t)$$

uniquely determines $\underline{V}_{t_0}(t)$ for a given $\underline{B}(t)$, one can replace κ in (3.15) by a functional κ such that

$$\begin{aligned} \underline{T}(x, t) &= \kappa \int_{\tau=t_0}^t (\underline{U}_t(\tau), \underline{V}_{t_0}(t)) \\ &= \kappa \int_{\tau=t_0}^t (\underline{B}(t), \underline{C}_t(\tau)). \end{aligned} \quad (9.1)$$

Without loss of generality, we can set $t_0 = -\infty$ in (9.1) to rewrite it in the more familiar form

$$\underline{T} = \kappa \int_{\tau=-\infty}^t (\underline{B}(t), \underline{C}_t(\tau)) \quad (9.2)$$

given by Truesdell and Noll [35]. We wish to seek a specific approximate representation of (9.2) for a generalization, under conditions of moderate strain rates, of the model in Fig. 2.3.

For this we imagine a homogeneous matrix of a viscoelastic solid with a stress free natural configuration κ exhibiting isotropic material symmetry. It is helpful to imagine that this matrix is made up of random distribution of elastic rod-like solids and flexible melt like macromolecules.

During any straining at a time $t \in [t_0, t]$, the relative rate of change of elastic stress $\dot{\underline{T}}_t(\tau)$ with respect to the current configuration at time t can be modeled as

$$\dot{\pi}_t(\tau) = \dot{\pi}_t^s(\tau) + \dot{\pi}_t^f(\tau), \quad (9.3)$$

where

$$\dot{\pi}_t^s(\tau) = \frac{d}{d\tau} [\phi_{C_t(t_0)}(C_t(\tau))] \quad (9.4)$$

is the same as in (4.5) and

$$\dot{\pi}_t^f(\tau) = \frac{d}{d\tau} [\phi(C_t(\tau))] \quad (9.5)$$

is the same as in (6.5). Furthermore, by analogy to the case in Fig. 2.3, we can postulate that the evolution of the Cauchy stress at any $\tau \in [t_0, t]$ is given as

$$\dot{T}(\xi, \tau) = \dot{T}_s(\xi, \tau) + \dot{T}_f(\xi, \tau), \quad (9.6)$$

where

$$\dot{T}_s(\xi, \tau) = \dot{\pi}_t^s(\xi, \tau) \quad (9.7)$$

and

$$\dot{T}_f(\xi, \tau) = \frac{d}{d\tau} \mathcal{R}_{\tau=t_0}^{\tau} \left[\frac{d}{d\eta} \phi(C_t(\eta)) \right]. \quad (9.8)$$

Now integration of (9.6) for an incompressible solid yields

$$T(x, t) = -pI + g^K(B(t)) + \mathcal{R}_{\tau=t_0}^t [\dot{\pi}_t^f(\tau)]. \quad (9.9)$$

The function g^K in (9.9) is as defined in (4.1). Let $t_0 \rightarrow -\infty$ and $B(t) = \lim_{t_0 \rightarrow -\infty} C_t(t_0)$ be such that the solid part of the matrix is in its elastic realm governed by (9.7). Following the approximation leading to (8.4), at moderate strain rates, T in (9.9) can be written as

$$T = -pI + g^K(B(t)) + \int_0^\infty (-G'(\sigma)) \{f_1(I(t-\sigma), II(t-\sigma)) C_t(t-\sigma) + f_2(I(t-\sigma), II(t-\sigma)) C_t^{-1}(t-\sigma)\} d\sigma, \quad (9.10)$$

where

$$g^K(B(t)) = B_1(I_1(t), I_2(t)) B(t) \quad (9.11)$$

$$+ \beta_2 (I_1)(t), I_2(t)) \tilde{B}^{-1}(t).$$

The last term in (9.10) is as in (8.4). The material functions β_1 and β_2 in (9.11) are functions of

$$I_1(t) \stackrel{\text{def}}{=} \text{tr} \tilde{B}(t), \quad (9.12)$$

and

$$I_2(t) \stackrel{\text{def}}{=} \text{tr} \tilde{B}^{-1}(t).$$

10. The generalization of Jeffrey's liquid, and Kelvin-Voigt Solid

For polymer solutions in a low molecular weight Newtonian solvent, one expects that the effect of the solvent is a rapid relaxation associated with the dashpot in Fig. 2.4. The first term in the right side of (2.8) can be shown to generalize to a Newtonian term

$$\mu \frac{d}{dt} (\epsilon(\tau) - \epsilon(t)) \Big|_{\tau=t} \rightarrow \mu \dot{\underline{C}}_t(\tau) \Big|_{\tau=t} \quad (10.1)$$

$$= 2\mu \underline{D}(\underline{x}, t).$$

In (10.1) above, \underline{D} is the symmetric part of the velocity gradient "grad $\underline{v}(\underline{x}, t)$ ". Using (10.1) and (8.4) it follows that (2.8) for an incompressible viscoelastic fluid may be generalized as

$$\underline{T} = p\underline{I} + 2\mu\underline{D} + \int_0^\infty (-G'(s)) \{f_1(I, II) \underline{C}_t(t-s) + f_2(I, II) \underline{C}_t^{-1}(t-s)\} ds. \quad (10.2)$$

It is well known that (10.2) also follows from the strictly viscoelastic model in (8.4) if we formally replace $G(s)$ by

$$\mu \delta(s-0) + G(s), \quad (10.3)$$

where $\delta(s-0)$ is the Dirac delta function and $\mu > 0$. A theoretical justification for allowing distributions in the kernel of the linearized constitutive relation has been discussed by Saut and Joseph [31]. It is conceivable that the so called *exact* Newtonian contribution in (10.3) is merely an idealized way of asserting a rapid relaxation present in a regular $G(s)$ near $s=0$.

Accepting the generalization in (10.1) and (4.7), a Kelvin-Voigt incompressible solid ([17], p. 21) generalizes to give

$$\underline{T} = -p\underline{I} + g^K(\underline{B}(t)) + 2\mu\underline{D}(\underline{x}, t). \quad (10.4)$$

Similarly, a parallel combination of a Kelvin-Voigt element and a Maxwell element generalizes to give

$$\underline{T} = g^K(\underline{B}) + \underline{T}_{\text{Jeffrey}},$$

(10.5)

where $\underline{T}_{\text{Jeffrey}}$ is given by the right side of (10.2).

11. Compatibility of the K-BKZ model with some successful semi-empirical models in shear flows.

Of particular significance is the fact that some successful semi-empirical models in shearing motion are implied by the K-BKZ model. Consider a simple shearing motion in y direction and in x-y plane; the motion $\underline{x} = \chi(\underline{X}, t)$ of a particle $\underline{X} (X, Y, Z)$ is given in Cartesian co-ordinates by the relation

$$(x, y, z) = (X, Y + \mu(x, t), Z). \quad (11.1)$$

The shear strain $\gamma(t)$ and the relative shear strain $\gamma_t(s)$ are defined as

$$\gamma(t) = \frac{\partial \mu}{\partial x}(x, t),$$

and

$$\gamma_t(s) = \frac{\partial \mu}{\partial x}(x, t-s) - \frac{\partial \mu}{\partial x}(x, t). \quad (11.2)$$

A standard calculation shows that the Cartesian components of $\underline{C}_t(s)$ is given by

$$[\underline{C}_t(t-s)] = [\underline{C}_t(s)] = \begin{bmatrix} 1 + \gamma_t(s)^2 & \gamma_t(s) & 0 \\ \gamma_t(s) & 1 & 0 \\ 0 & 0 & 1 \end{bmatrix} \quad (11.3)$$

The corresponding components of $\underline{C}_t^{-1}(t-s)$ is given by

$$[\underline{C}_t^{-1}(t-s)] = \begin{bmatrix} 1 & -\gamma_t(s) & 0 \\ -\gamma_t(s) & 1 + \gamma_t^2(s) & 0 \\ 0 & 0 & 1 \end{bmatrix} \quad (11.4)$$

From (11.3) and (11.4), it follows that

$$I = \text{tr } \underline{C}_t(t-s) = 3 + \gamma_t(s)^2,$$

and

$$(11.5)$$

$$II = \text{tr } C_t^{-1}(t-s) = 3 + \gamma_t(s)^2.$$

Substituting (11.5) in (8.4), we find that the shear stress $T_{\langle xy \rangle}$ is given by

$$T_{\langle xy \rangle} = \int_0^\infty (-G'(s)) \psi(\gamma_t^2(s)) \{-\gamma_t(s)\} ds, \quad (11.6)$$

where

$$\psi(\gamma_t^2(s)) \stackrel{\text{def}}{=} f_2(3+\gamma_t(s)^2, 3+\gamma_t(s)^2) - f_1(3+\gamma_t(s)^2, 3+\gamma_t(s)^2).$$

Similarly the first normal stress difference $N_1 \stackrel{\text{def}}{=} T_{\langle yy \rangle} - T_{\langle xx \rangle}$ and the second normal stress difference $N_2 \stackrel{\text{def}}{=} T_{\langle xx \rangle} - T_{\langle zz \rangle}$ are given by

$$N_1 = \int_0^\infty (-G'(\sigma)) \psi(\gamma_t^2(\sigma)) \gamma_t(\sigma)^2 d\sigma, \quad (11.7)$$

and

$$N_2 = \int_0^\infty (-G'(\sigma)) f_1(3+\gamma_t^2(\sigma), 3+\gamma_t^2(\sigma)) \gamma_t^2(\sigma) d\sigma.$$

The function G and ψ in (11.6) and (11.7) are chosen by Laun [20] to agree with experimental data obtained from measurements of steady viscosity and transient response to step-strain. In Narain and Joseph [24], it was shown that the data of step-strain experiments are reliable only at times larger than a characteristic relaxation time. For a fluid confined between two flat parallel plates, one of the plates is given a sudden displacement, and if the effects of transients die quickly then the assumption of step-strain gives useful asymptotic results [24]. Therefore if

$$\frac{\partial \mu}{\partial x}(x, t) = \begin{cases} 1, & \text{for } t > 0 \\ 0 & \text{for } t < 0. \end{cases} \quad (11.8)$$

we get

$$\gamma_t(s) = \begin{cases} 0 & \text{for } 0 < s < t \\ -\gamma_0 & \text{for } s \geq t \end{cases} \quad (11.9)$$

$$\stackrel{\text{def}}{=} -\gamma_0 [1 - H(t-s)]$$

Substitution of (11.8) and (11.9) in (11.6) and (11.7), one arrives at

$$T^{<xy>} = \int_t^\infty (-G'(s)) \psi(\gamma_0^2) \gamma_0 ds, \quad (11.10)$$

and

$$N_1 = \int_t^\infty (-G'(s)) \psi(\gamma_0^2) \gamma_0^2 ds.$$

Now (11.10) yields the remarkable result

$$T^{<xy>}(t)/\gamma_0 = N_1(t)/\gamma_0^2. \quad (11.11)$$

Eqs. (11.10) and (11.11) are in agreement with the semi-empirical formulas (15) and (16) of Laun's paper [20]. The modeling for low density polyethylene was done with the following choices of G and ψ :

$$G(s) = \sum a_i \exp\left(-\frac{s}{\tau_i}\right) \quad (11.12)$$

and

$$\psi(\gamma_t^2(s)) = \bar{f}_1 \exp(-n_1 |\gamma_t(s)|) + \bar{f}_2 \exp(-n_2 |\gamma_t(s)|).$$

In (11.12); τ_i , $i = 1, 2, \dots$ is interpreted as a discretization of a continuous spectrum ([17], p. 81) of relaxation times and τ_i depends on temperature T through the Arrhenius type relation ([17], p. 73)

$$\tau_i(T) = \exp \left[\frac{E_0}{R} \left(\frac{1}{T} - \frac{1}{T_0} \right) \right] \tau_i(T_0). \quad (11.13)$$

For the choice of constants in (11.12) and (11.13), the reader is referred to Laun [20]. The remarkable validity of eq. (11.11) has been noted by Wagner ([37], [38]), and Laun [20]. This validity of (11.11) for a low density polyethylene melt is shown in Fig. 11.1.

We also note that step in shear rate, start up of elongational flows, and shear dependent steady viscosities for melts can all be modeled (see [19]) by (8.4). Successful modelings done by Wagner [36, 37], Chang and Lodge [7], and Laun [20] can be reinterpreted in the context of the K-BKZ model. The superiority of this model in various flow regimes of moderate strain rates has already been emphasized by Tanner [33].

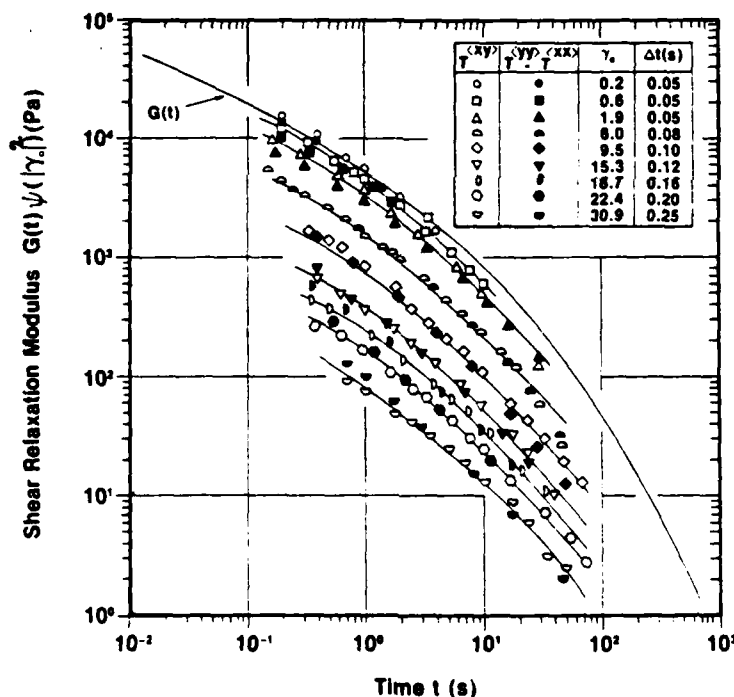


Fig. 11.1: In the above figure the shear relaxation modulus $G(t)\psi(\gamma_0^2)$ is computed through (11.10)₁ as $T^{<xy>}/\gamma_0$ and through (11.10)₂ as $T^{<yy>} - T^{<xx>}/\gamma_0^2$. In the figure Δt is the approximate rise time. The results are taken from Fig. 5 of Laun [20]. The experiments were done at $T = 150^\circ\text{C}$ and different shear strains γ_0 . For $t < 1$ sec, the data is not reliable because of the effects of transients.

APPENDIX

A) Let us assume the kinematics of section 3 associated with configuration κ and write

$$\tilde{\xi} = {}^{\kappa}\chi_{t_0}(\tilde{x}, \tau), \quad (A.1)$$

where

$$\kappa(P) = \tilde{x}, \quad t_0 \leq \tau \leq t.$$

The constitutive theory leading to (3.18) assumes that the material is undistorted up to time t_0 . But this means that any other undistorted configuration μ of the particle P at a time t_0 could at most be a rigid rotation of the configuration κ . To describe this, let

$$\kappa(P) = \tilde{x}, \quad (A.2)$$

and

$$\mu(P) = y.$$

Let λ be the invertible function which maps κ onto μ such that

$$y = \lambda(\tilde{x}). \quad (A.3)$$

Now, for $t_0 \leq \tau \leq t$, we introduce

$$\begin{aligned} \tilde{\xi} &= {}^{\kappa}\chi_{t_0}(\lambda^{-1}(y), \tau) \\ &\stackrel{\text{def}}{=} {}^{\mu}\chi_{t_0}(y, \tau). \end{aligned} \quad (A.4)$$

From (A.2) and (A.3):

$${}^{\mu}\tilde{F}_{t_0}(\tau) = {}^{\kappa}\tilde{F}_{t_0}(\tau) \nabla \lambda, \quad (A.5)$$

for $\tau \in [t_0, t]$. In (A.4); ${}^{\mu}\tilde{F}_{t_0}(\tau)$, ${}^{\kappa}\tilde{F}_{t_0}(\tau)$ and $\nabla \lambda$ are respective gradients of ${}^{\mu}\chi_{t_0}(y, \tau)$, ${}^{\kappa}\chi_{t_0}(\tilde{x}, \tau)$, and λ . The relation in (A.5) implies that the two configurations (not pairs of par-

tics) κ and μ undergo the same changes in shape-history. The two configurations κ and μ are undistorted configurations of the same material element only if

$$\nabla \lambda \in \text{Orth} \quad (\text{A.6})$$

is an orthogonal tensor. The orthogonality of $\nabla \lambda$ in (A.6) yields the result

$${}^{\kappa} \underline{U}_t(\tau) = {}^{\mu} \underline{U}_t(\tau) \quad (\text{A.7})$$

for all $t_0 \leq \tau \leq t$. Now the natural definition of the response functionals ${}^{\kappa} \mathcal{H}$ and ${}^{\mu} \mathcal{H}$ (as introduced in (3.12)) are:

$$\begin{aligned} \tilde{T} &= {}^{\kappa} \mathcal{H} \left({}^{\kappa} \underline{U}_t(\tau), {}^{\kappa} \underline{F}_{t_0}(t) \right) \\ &= {}^{\mu} \mathcal{H} \left({}^{\mu} \underline{U}_t(\tau), {}^{\mu} \underline{F}_{t_0}(t) \right). \end{aligned} \quad (\text{A.8})$$

On substituting (A.5) and (A.7) in (A.8)₁ we find

$$\tilde{T} = {}^{\kappa} \mathcal{H} \left({}^{\mu} \underline{U}_t(\tau), {}^{\mu} \underline{F}_{t_0}(t) (\nabla \lambda)^{-1} \right). \quad (\text{A.9})$$

Since $(\nabla \lambda)^{-1}$ in (A.9) is orthogonal, use of the definition (3.14) of isotropic material symmetry of the configuration κ implies that

$$\begin{aligned} \tilde{T} &= {}^{\kappa} \mathcal{H} \left({}^{\mu} \underline{U}_t(\tau), {}^{\mu} \underline{F}_{t_0}(t) (\nabla \lambda)^{-1} \right) \\ &= {}^{\kappa} \mathcal{H} \left({}^{\mu} \underline{U}_t(\tau), {}^{\mu} \underline{F}_{t_0}(t) \right). \end{aligned} \quad (\text{A.10})$$

Therefore, for an isotropic simple material, combining (A.8) and (A.10), we have

$$\tilde{T} = {}^{\kappa} \mathcal{H} \left({}^{\mu} \underline{U}_t(\tau), {}^{\mu} \underline{F}_{t_0}(t) \right) \quad (\text{A.11})$$

$$= {}^{\mu} \int_{t_0}^t (\mu_{\underline{U}_t(\tau)}, \mu_{\underline{F}_{t_0}(t)}).$$

The result above establishes that for an isotropic simple material, two undistorted real states have the same representation for their response functionals. Now if the arguments given through (3.12) to (3.18) are repeated, we find

$$\begin{aligned} \underline{T} &= {}^{\kappa} \int_{\tau=t_0}^t [\underline{C}_t(\tau), \underline{C}_t(t_0)] \\ &= {}^{\mu} \int_{\tau=t_0}^t [\underline{C}_t(\tau), \underline{C}_t(t_0)], \end{aligned} \quad (\text{A.12})$$

where

$${}^{\kappa} \underline{C}_t(\tau) = {}^{\mu} \underline{C}_t(\tau) \stackrel{\text{def}}{=} \underline{C}_t(\tau)$$

for $t_0 \leq \tau \leq t$. Furthermore, if the material is undistorted up to time t_0 , we postulate that the representation of ${}^{\kappa} \underline{y}$ in (3.18) must be such that

$$\begin{aligned} \underline{T} &= {}^{\kappa} \int_{\tau=t_0}^t [\underline{C}_t(\tau), \underline{C}_t(t_0)] \\ &= {}^{\kappa} \int_{\tau=t_*}^t [\underline{C}_t(\tau), \underline{C}_t(t_*)] \end{aligned} \quad (\text{A.13})$$

for any $t_* \leq t_0$. The motivation for (A.13) is the simple undistorted rest state ${}^{\kappa} \chi_{t_0}(\underline{X}, \tau) = \underline{X}$ for all $\tau \leq t_0$.

B) For an elastic solid, (3.5) reduces to

$$\begin{aligned} \underline{T}(\underline{x}, t) &= {}^{\kappa} \int_{\tau=t_0}^t (\underline{F}_{t_0}(\tau)) \\ &= {}^{\kappa} \phi_1(\underline{F}_{t_0}(t)), \end{aligned} \quad (\text{B.1})$$

and therefore

$$\underline{T}(\underline{\xi}, \tau) = {}^{\kappa} \phi_1(\underline{F}_{t_0}(\tau)).$$

In (B.1), ${}^{\kappa} \phi_1 : \mathcal{J} \rightarrow \mathcal{J}^{\text{sym}}$ such that ${}^{\kappa} \phi_1(1) = 0$. If the current configuration is used with the help of (3.6), we can define

$$\begin{aligned}\underline{T}(\underline{\xi}, \tau) &= {}^K\phi_1(\underline{F}_t(\tau), \underline{F}_{t_0}(t)) \\ &\equiv {}^K\phi_2(\underline{F}_t(\tau), \underline{F}_{t_0}(t)),\end{aligned}\quad (B.2)$$

where ${}^K\phi_2: \mathcal{V} \times \mathcal{V} \rightarrow \mathcal{V}^{\text{sym}}$. Furthermore, if the arguments leading to (3.18) are repeated for the special case of isotropic elastic solid given by (B.1) and (B.2), it is easily seen that there is a response function ${}^K\phi$ such that

$$\underline{T}(\underline{\xi}, \tau) = {}^K\phi(\underline{C}_t(\tau), \underline{C}_t(t_0)), \quad (B.3)$$

and it has the obvious values

$$\begin{aligned}\underline{T}(\underline{x}, t_0) &= {}^K\phi(\underline{C}_t(t_0), \underline{C}_t(t_0)) \\ &= \underline{0},\end{aligned}$$

and

$$\underline{T}(\underline{x}, t) = {}^K\phi(\underline{1}, \underline{C}_t(t_0)).$$

The result in (A.11) of Appendix A emphasizes that the choice of any other undistorted configuration will yield the same representation of ${}^K\phi$ in (B.3). We use this to assert that there is a response function $\phi_{\underline{C}_t(t_0)}$ (determined by the natural state of the elastic solid) such that the Cauchy stress at a time $\tau \in [t_0, t]$ is given by

$$\underline{T}(\underline{\xi}, \tau) = \phi_{\underline{C}_t(t_0)}(\underline{C}_t(\tau)). \quad (B.4)$$

Furthermore, in this special case, the Objectivity requirement in (3.20) reduces to

$$\underline{\Phi} \underline{Q} \underline{C}_t(t_0) \underline{Q}^T (\underline{Q} \underline{C}_t(\tau) \underline{Q}^T) = \underline{Q} [\phi_{\underline{C}_t(t_0)}(\underline{C}_t(\tau))] \underline{Q}^T \quad (B.5)$$

for every orthogonal tensor \underline{Q} .

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20. ABSTRACT (Continue on reverse side if necessary and identify by block number) An alternate constitutive formulation for visco-elastic materials, with particular emphasis on macromolecular viscoelastic fluids, is presented by generalizing Maxwell's idealized separation of elastic and relaxation mechanisms. The notion of relative rate of change of elastic stress is identified, abstracted, and formulated with the help of the established theory of finitely elastic isotropic materials. This gives a local rate type constitutive relation for an elastic mechanism in a simple material. For the simplest class of viscoelastic polymer melts, the notion of rate		

ABSTRACT (continued)

of change of elastic stress and its damped accumulation is identified and formulated. Under conditions of moderate strain rates, this scheme implies the reliable K-BKZ model for a class of polymer melts. An obvious extension generalizes the remaining classical spring-dashpot models.

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